Adatom condensation and quantum dot sizes in InGaAs/GaAs (001)

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The sizes and concentrations of capped and surface InGaAs/GaAs quantum dots (QDs) grown under the same conditions have been investigated. Comparisons obtained with transmission electron microscopy and scanning probe microscopy imaging show a significant enlargement in the sizes of surface QDs compared with capped QDs. This discrepancy in dot dimensions increases with decreasing island surface densities and can be partially explained by thermal adatom condensation during sample cooling. These findings suggest a technique to estimate adatom concentrations and their migration lengths in strained heteroepitaxy. © *2000 American Institute of Physics.* [S0003-6951(00)03312-X]

Accurate resolution of the sizes of quantum dots (QDs) is important for most of their device applications, since QDs emission energies, number of excited states, energy sublevels, charging energies, Coulomb interactions, and other optoelectronic properties are determined by their confinement dimensions. The interest in developing nanotechnology using Stranski–Krastanow (S–K) QDs makes structural characterization an important component of research in nanostructures. The scanning probe technique of atomic force microscopy (AFM) is often used to estimate sizes of dots before capping. The implicit assumption is that if all growth conditions are the same, the size of the capped dots which produce the luminescence emission is the same as the uncapped dots as characterized using AFM. Transmission electron microscopy (TEM) can be used to examine the structure of QDs after capping, and capped dots are the most relevant since they produce the optical/electronic signals of interest in device applications. However, TEM is harder to use and not always available to researchers investigating QDs for device applications. Establishing a correspondence between capped and uncapped QD structures is therefore of interest.

Here we compare sizes of InGaAs islands before and after these are capped with the GaAs barrier. We find significant differences in the sizes between capped and uncapped islands, and this size discrepancy is an inverse function of QD surface densities. Large variations in InGaAs QD concentrations were obtained with two different methods: (i) by simultaneous growths on vicinal GaAs [001] substrates with a range in surface step densities obtained by slight variations in substrate miscut angles;[1] and (ii) by using a graded growth rate which results in a positionally varying QD density over a few millimeters.[2] The increase in size of uncapped islands upon cooling can be qualitatively explained by group III thermal adatom condensation on surface islands.

The following information applies to all structures. InGaAs/GaAs QDs were grown by metalorganic chemical vapor deposition (MOCVD). $(CH_3)_3Ga$, $(CH_3)_3In$, and AsH_3 were used as precursors. After growth of GaAs buffer layers at 650 °C, the temperature was lowered to 550 °C and nanometer sized InGaAs islands were grown by depositing 5 ML with nominal composition $In_{0.6}Ga_{0.4}As$. In the first type of growth, the H_2 carrier flow rate was 17.5 standard liters per minute (slm) to achieve uniform growth, allowing comparisons of island statistics on multiple substrates with one deposition. Different QD densities were obtained on (001) GaAs substrates with slight variations in miscut angle (θ_m):0.00°, 0.25°, 0.50°, 0.75°, 1.25°, and 2° (all $\pm 0.25°$) towards (110). GaAs capping layers were 100 nm when used. In the second type of growth, graded growth rates (from 0.5 to 0.75 ML/s) were obtained using a H_2 carrier flow rate of 5 slm on GaAs (001). These graded structures were grown for two different values of arsine partial pressures (5.7×10^{-2} and 2.2×10^{-1}). When used, the GaAs capping layer thickness was 30 nm (not graded). After formation of the QDs, the flow of group III sources was interrupted but the arsine flow was kept while the structures cooled down to below 400 °C. All precursor gases were removed as the samples cooled from 400 °C to room temperature. Island sizes, shapes, and concentrations were measured using frequency modulation (FM) (in contact mode with etched silicon nitride tips) and plan view TEM. TEM specimens were first mechanically thinned to ~100 μm and locally thinned to ~50 μm with a dimple/grinder. The final thinning was accomplished with a bromine/methanol etch, or with a solution of H_2SO_4:H_2O_2:H_2O (8:1:1). Philips EM430 (300 keV) and CM12 (120 keV) TEMs were used to obtain on zone bright field images of both capped and uncapped QDs.

Figure 1(a) shows the variation in QD concentration by miscut angle variation (first type of growth described earlier). Steps are energetically favorable sites for island nucleation; therefore, large variations in QD concentrations can be obtained by varying the availability of surface steps with growth conditions that minimize island coverage.[1] Figure 1(b) shows size differences between surface dots and capped dots determined by FM and plan view TEM imaging, respectively, indicating that surface dots are larger, and that this size discrepancy increases with decreasing QD surface density.

Figures 2 and 3 show the results from the second type of

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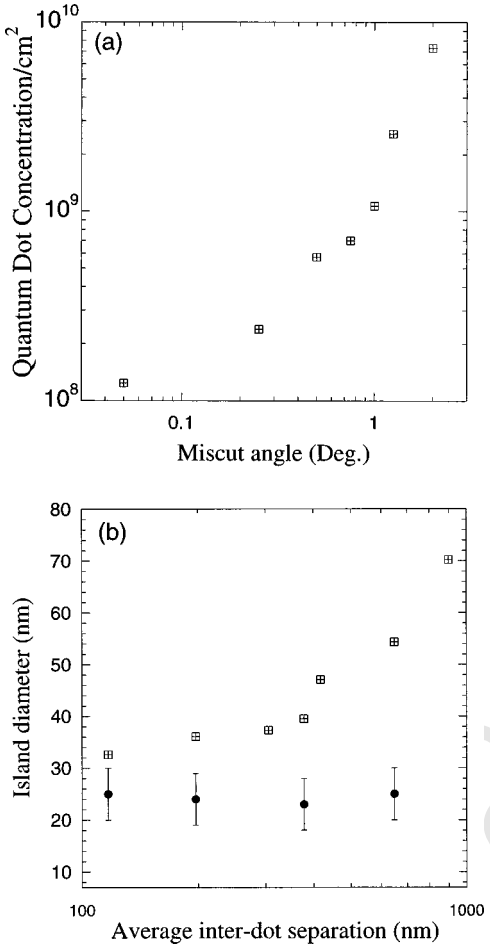

FIG. 1. (a) Variation in QD concentration as a function of substrate miscut angle in simultaneous growths at 550 °C. Measurements between capped and uncapped islands show good correspondence. (b) Variations in average dot diameter for surface (crossed squares) and capped (filled circles) islands as a function of average dot–dot separation for the same growth, obtained by FM and plan view TEM, respectively.

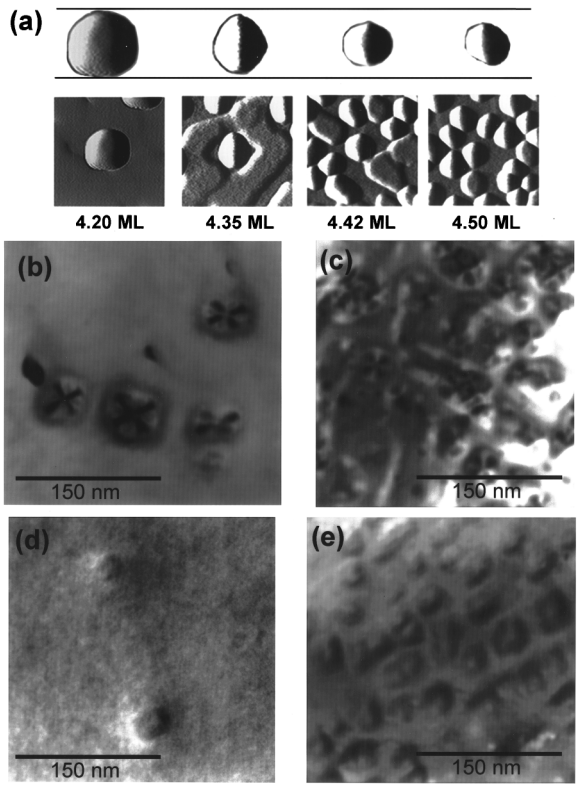

FIG. 2. (a) Composite FM deflection image showing island size evolution from low island concentration to island saturation. Above each 250 nm $\times$250 nm frame is a magnified image of a typical island. (b) and (c) plan view on-zone bright field TEM images of uncapped; (d) and (e) ~~and~~ capped InGaAs/GaAs QDs.

growth (using conditions that produce a graded growth rate) which allows a study to be made of the entire growth evolution of S–K QDs with a single deposition.[2] Saturation island densities can be maximized at low arsine partial pressure and the resulting QDs are stable against ripening even after prolonged annealing in arsine. At higher values of arsine partial pressure, saturation island densities are reduced (from low 10^{10} to mid 10^9 islands/cm^2). Both low and high arsine flows were used in these graded structures, to evaluate possible effects from this important parameter. The plots of island concentration as a function of ML deposition[3] shown in Fig. 3(a) indicate that the critical thickness for the S–K transformation depends on arsine partial pressure. Figure 3(b) shows differences in island sizes obtained from surface islands and from capped islands grown under the same conditions. Surface islands were characterized by both TEM and FM, to account for possible technique related effects. Figure 3(b) also shows a general trend of increasing island sizes with decreasing island concentrations; however, this increase seems to saturate for islands obtained with deposition at low arsine partial pressures.

We report differences in capped and uncapped island sizes observed from different growth experiments. Surface island size estimates also agree across both types of structural techniques, therefore, we believe these differences to be real despite potential errors from technique related artifacts. Surface islands, mainly for low concentrations, are significantly larger than their capped counterparts. It should also be noted that there is good agreement between concentration values determined from capped and uncapped islands. The latter observation eliminates Ostwald ripening during sample cooling as a major contributor to this size discrepancy.

The origin for this island size discrepancy will now be examined. We suggest that a common mechanism takes place in both types of deposition described. The island size difference can be explained by group III thermal adatom condensation on existing islands during sample cooling. As shown in recent studies after scanning tunneling microscopy analysis of rapidly quenched GaAs surfaces,[4,5] Ga adatom concentrations on GaAs (001) surfaces are high (a significant fraction of a ML) at the growth temperatures normally used in molecular beam epitaxy and MOCVD growth. This indicates that growth on GaAs (100) is closer to equilibrium than previously assumed. This observation can also explain the island size differences presented here. The islands act as a ''sink'' or ''seed'' for condensing adatoms, which migrate from the surface area surrounding each island. Since cooling is slow, adatom diffusion still permits migration to existing islands for small and intermediate interisland separations. For widely spaced islands, migration lengths become a limitation, and this can explain the saturation in surface island dimensions seen in one of the evolution growths shown in Fig. 3(b). Another obvious sink for condensing adatoms are surface steps, and the relative importance of each type of

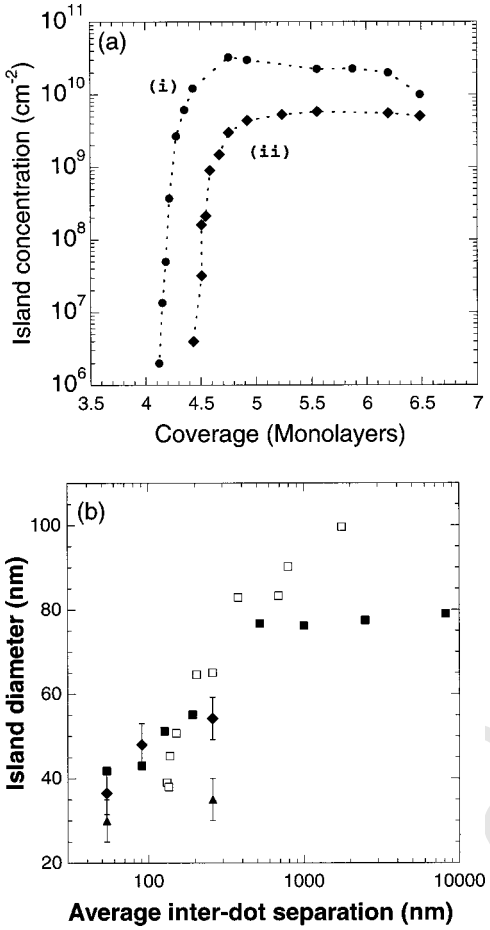

FIG. 3. (a) Variations in InGaAs island concentrations as a function of ML coverage obtained for graded structures with a positionally varying growth rate (see Ref. 2) for low (i) and high (ii) values of arsine partial pressure (5.7×10^{-2} and 2.2×10^{-1}, respectively). (b) Island diameters as a function of average interdot separations for graded depositions used in (a). All filled symbols represent data for the growth at low arsine partial pressure. The squares are surface dots measured by AFM, diamonds are also surface dots measured by plan view TEM, and triangles are capped dots (plan view TEM). Unfilled squares correspond to the islands formed at high arsine partial pressures.

sink (islands or steps) will depend on the island surface concentration, and average step separation (350 nm for the surfaces used in the island ''evolution'' experiments). For average dot separations of the order of the step separations, adatoms will condense on both islands and steps, but for interisland separations $\ll$ step separation, most adatom will condense on islands, given sufficiently long migration lengths.

A relative estimate for adatom concentration could then be made from the results shown in Fig. 3. If we consider that before cooling the islands had the same size as the capped QDs (there are no significant interdiffusion effects in this material system at the growth temperatures used), then the condensing adatoms contribute an increase in volume equal to the uncapped island size minus the capped island size. If we assume that migrating adatoms will condense on each island, the contributing surface will be distributed over a square area of side equal to the average interdot separation, minus the area occupied by the uncapped dot. Using reasonable values for seed island sizes (within the range of capped island sizes) and using thermal adatom concentrations obtained in Ref. 4, the growth of islands at intermediate concentrations can be accounted by condensing adatoms. However, the low concentration islands do not show sufficient volume differences to account for all surrounding adatoms. In other words, if all adatoms condensed on the low density islands, these would be even larger. The last observation can easily be explained by competing sinks for adatoms as the islands become more widely separated. An accurate value for adatom concentration cannot be obtained without knowing the exact size of the capped seed islands,[6,7] but if we assume a certain fixed size for the seed islands, we can confirm that the condensing adatom contribution is greater (a factor of 2–3) with the growths done at low values of arsine partial pressures than for the growths done at high values of partial pressure. This is expected since raising the As pressure will raise this component's chemical potential and therefore lower the Ga (and In) chemical potential and hence the Ga (and In) adatom density.[4,5]

The differences shown in Fig. 3(b) for the island evolution growths done at different values of arsine partial pressures indicate that adatom migration lengths are lower for growths done at low values of arsine partial pressures. Diffusion lengths can be roughly estimated from the threshold concentration for which island sizes cease to increase, giving 1/4–1/3 μm for the growth done at low arsine partial pressures as, and $\geqslant 1$ μm for higher arsine partial pressures. It is not possible to determine for which group III adatom this corresponds to, but a weighted average is a reasonable assumption. This can be investigated further with the growth of binary islands. Performing similar experiments with InAs QDs will also eliminate possible effects from indium enrichment in ternary alloy[8,9] since differences in indium enrichment could induce local differences in island ternary composition, which could affect island size distributions.

In conclusion, we have shown that the size of surface and capped InGaAs QDs differ, and that this discrepancy increases with decreasing island coverage. Differences in island coverage were obtained with simultaneous growths on different GaAs (100) vicinal surfaces and with structures deposited at growth rates varying with position. We explain these size differences by condensing adatoms from different surface areas surrounding low and high-density islands.

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